A study of the linearity of transfer leaks and a helium leak detector

C. D. Ehrlich, S. A. Tison, H. Y. Hsiao,[a] and D. B. Ward
Thermophysics Division, National Institute of Standards and Technology, Gaithersburg, Maryland 20899

(Received 30 April 1990; accepted 21 July 1990)

A study has been performed of the linearity of two types of variable-reservoir-pressure leaks and a commercial tuned magnetic sector mass-spectrometer helium leak detector. While the leaks exhibit predictable (but not always linear) behavior over a broad range, the linearity of the leak detector depends strongly on properly correcting for observed drift and random fluctuations in the measured leak rate.

I. INTRODUCTION

Fixed-reservoir helium permeation leaks are frequently used for one-point calibration of leak detectors. However, if the leak detector is nonlinear its calibrated accuracy will deteriorate when leak rates differ significantly from the point at which it was calibrated. Performing a multiple-point calibration using several leaks can improve calibration accuracy but is usually inconvenient, and requires a larger inventory of leaks. A technique needing a much smaller leak inventory may be based on a single, calibrated leak in which the pressure or concentration of gas in the reservoir may be conveniently varied. Such a leak will subsequently be referred to as a "variable" leak, or VL. A VL will preferrably have the property of a linear leak rate dependence on reservoir pressure or concentration over the desired range of use. We have used such a technique in this study to characterize the linearity of a leak detector that is part of the National Institute of Standards and Technology (NIST) Leak Calibration Service. The leak detector is used to characterize the temperature dependence of the leak rate from customer helium permeation leaks, which may change by an order of magnitude over the usual temperature calibration range of 50 °C. We examined two types of leak elements, sintered and helium permeation, to determine their suitability as transfer standards in determining the linearity of the leak detector. A helium permeation type VL was chosen due to its stability and linearity over the desired range of leak rates. The leak rate (referenced to a single temperature) of the variable-reservoir helium leak was measured as a function of the reservoir concentraion using the NIST Primary Leak Standard. This VL (NBSL40) was characterized with reservoir concentrations between 4×10^{-5} and 4.6×10^{-4} mol/cm^3, corresponding to reservoir pressures between 0.1 and 1.1 MPa, and to leak rates at 23 °C of 1.6×10^{-12}–1.9×10^{-11} mol/s. The VL was then used to determine the linearity of the leak detector, which utilizes a magnetic sector mass-spectrometer tuned for helium detection. We will show that, over more than a decade of reservoir concentration, the equilibrium leak rate of the helium permeation VL that we used is, to within experimental error, directly proportional to the concentration of the helium in the reservoir. This is consistent with permeation theory, which states that the equilibrium diffusion rate is a function of the concentration gradient, and not the pressure gradient, across the permeation element, as discussed in Sec. III B. Our preference for specifying reservoir concentration rather than reservoir pressure for helium permeation leaks is based on this observation. The leak detector was then calibrated over the range of the characterized VL, and from this data the linearity of the leak detector was determined.

II. DESCRIPTION OF APPARATUS

A. Primary leak standard

The primary leak standard that has been developed at NIST and used to determine the linearity of the VL is shown schematically in Fig. 1. The standard is divided into three major components: the vacuum chamber, the leak manifold, and the flowmeter. The flowmeter is described in detail in Ref. 1 and the vacuum chamber is similar to that described in Ref. 2. The leak manifold includes vacuum pumps for evacuation of the downstream side of the leaks in order to establish an equilibrium concentration gradient across the helium permeation leak elements prior to calibration; the leak manifold also includes pressure control systems to establish the reservoir pressure of VLs, and valving to connect different leaks to the primary standard.

To calibrate a leak on the NIST Primary Leak Standard, the leak is placed on the manifold, and the entire system is evacuated. Gas from the unknown leak is valved into the vacuum chamber, flows through an orifice, and is then evacuated by the pump. After the gas flow and upper chamber pressure reach equilibrium, the upper chamber helium partial pressure indication, as measured using a residual gas analyzer tuned to helium, is recorded. The leak is then isolated from the system, the flowmeter is pressurized with helium, and the flow is adjusted such that, upon stabilization, the upper chamber helium partial pressure indication is the same as the partial pressure recorded while the leak was connected to the vacuum chamber. The flow rate is then measured. Within limits determined primarily by instabilities of the upper chamber partial pressure gage, this measured flow rate can be equated to the flow rate of the unknown leak, and the calibration is complete. The uncertainties of the measured leak rate include a systematic uncertainty that is not greater than 7% (Ref. 1) and typical random uncertainties of 0.5%–2%, representing day-to-day fluctuations which may be a function of leak rate.

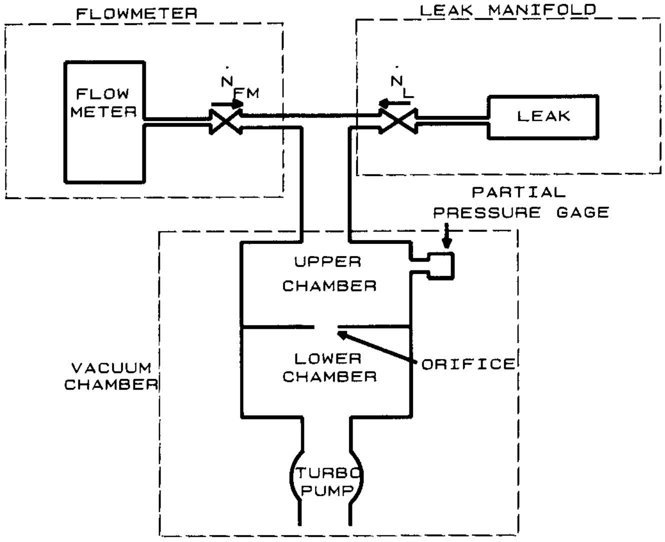

FIG. 1. Schematic of the NIST Primary Leak Standard.

B. Leak comparison system

The leak detector to be investigated is an integral part of the Leak Comparison System used at NIST (Ref. 3) to measure the temperature dependence of leaks. This system was originally designed and provided by the Sandia Primary Standards Laboratory; it was subsequently modified at NIST. The leak comparison system is composed of three main components: the leak detector, the leak manifold, and the control systems. The leak manifold provides a way of introducing gas flow from two leaks into the leak detector, either simultaneously or individually, without requiring venting of the system. The control system operates the valving and controls the temperature of the leak under test. Leak rates of helium permeation leaks generally increase by about a factor of 10 as the temperature is increased from 0 to 50 °C, which corresponds to the temperature range over which leaks are calibrated as part of the NIST Leak Calibration Service. Therefore knowledge of the linearity of the leak detector is of primary interest over at least a decade of leak rate variation.

The commercial leak detector uses a magnetic-sector mass spectrometer tuned to helium. The mass spectrometer consists of an ion source, a magnet, and an ion collector. Gas entering the mass spectrometer is ionized and then separated by its mass/charge ratio so that only helium ions strike the collector. The signal originating from the collector is then amplified and registered as a leak rate using a digital voltmeter and an automated data acquisition system.

III. LEAK ARTIFACT SELECTION AND MODELING

Two types of leak elements, permeation and sintered, were investigated to determine their adequacy for use as transfer standards in the determination of the linearity of the helium leak detector. To be used for this purpose, the leaks should demonstrate high repeatability and the leak rate should be variable over at least a decade by changing the reservoir helium concentration. It is also desirable that the measured variation of the leak rate with helium reservoir concentration be consistent with well known physical processes governing the flow of the helium and, for users without access to a primary leak standard, it is highly desirable that the leak rate be linear with the measured helium reservoir concentration.

A. Sintered leaks

The first type of leak element considered was the sintered type, and two were examined for their suitability for this application. The two sintered VLs used different sintered materials; the first (denoted leak No. 49) was stainless steel (commercially available) and was mounted in a steel tube, while the second (denoted NBSL5) was composed of silicon carbide and was mounted in a glass tube. Although the elements are different in composition, the fundamental properties of the two types should be the same. Flow through a sintered material can be considered as flow through a series of capillaries which may join and diverge throughout the sintered material. At low reservoir pressures where the mean free path of the helium atoms is comparable with or larger than the characteristic average void dimension of the sintered material, the physical process governing the flow should be molecular; in this case the flow rate would be linearly dependent on reservoir helium concentration or pressure. At higher reservoir pressures atomic interactions become significant; in this case the flow would be transitional or viscous, and nonlinear with reservoir concentration or pressure.

Experimental results shown in Figs. 2 and 3 indicate that for helium the flow through the sintered material remained viscous for helium reservoir pressures down to about 100 kPa, below which the flow entered the transition region for the metal element. This is seen in Fig. 2, where the slope of the 'conductance' versus pressure curve decreases in this region. Data, when expanded, in Fig. 2 indicate that the flow is molecular for pressures below 1 kPa, while corresponding data in Fig. 3 indicates that the flow remains viscous at this pressure. The nitrogen and argon data in Fig. 2 show the

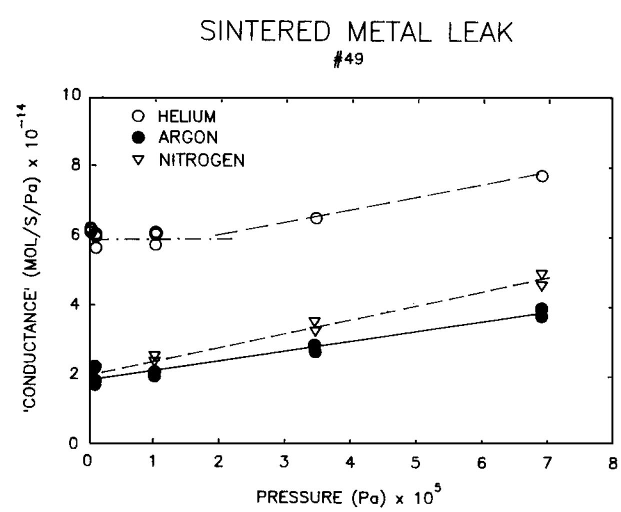

FIG. 2. Conductance of sintered metal leak vs reservoir pressure.

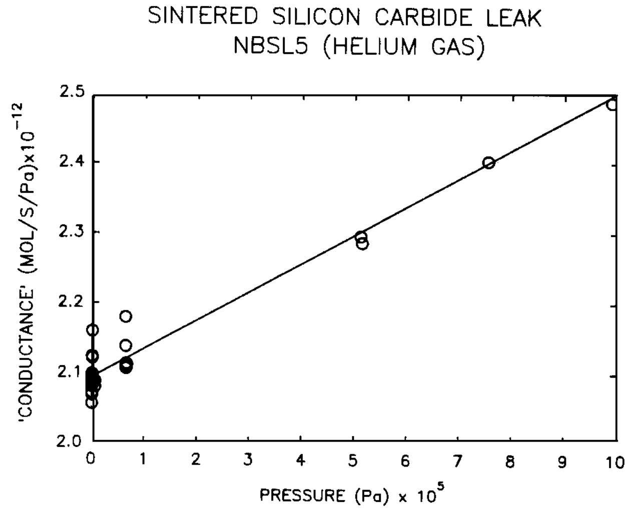

FIG. 3. Conductance of sintered silicon carbide leak vs reservoir pressure.

onset of transition flow at lower reservoir pressures than for helium, as expected. Since the onset of transition flow is governed by the characteristic length of the sintered material, any partial or total plugging of pathways would change the pressure at which transition occurs. This plugging could lead to significant changes in the leak rate for a given helium reservoir concentration. Furthermore, any changes in the structure of the sintered material caused by pressure or temperature cycling, or other events, could have the same effect. Past data have shown that metal sintered leaks operating in the low-flow-rate regime of interest in this study may exibit significant shifts, on the order of 15%, in a 3-month period.[3]

B. Helium permeation leaks

Permeation leaks emit helium due to the permeation or diffusion of the helium through the glass element caused by concentration gradients of helium atoms within the glass. For "reasonable" concentration gradients, the time-averaged flux density of gas at some point $\mathbf{r}$ in the glass, denoted $\mathbf{J}(\mathbf{r})$, is related to the local concentration in the glass, denoted $n(\mathbf{r})$, by Fick's diffusion equation:[4]

$$\mathbf{J}(\mathbf{r}) = -D\cdot\mathbf{grad}[n(\mathbf{r})], \quad (1)$$

where D is referred to as the diffusivity, or diffusion coefficient, and **grad** is the vector gradient operator.

Fick's law states that the rate of flux of a species is directly proportional to the gradient of the species concentration. At low gas pressures the dissolved helium occupies a very small percentage of the available interstices and it is reasonable to assume that the diffusion process obeys the simple linear form of Fick's law. However, Shelby showed in Refs. 5 and 6 that for vitreous SiO_2 as many as 50% of the available interstices may be occupied at pressures of 60 MPa (600 atm). Under these conditions gas migration becomes more complex, since the effects of interactions between diffusing atoms become important. At high pressures departure from the ideal gas law must also be taken into account and the activity coefficient for the gas is no longer unity; in this case the pressure must be replaced by fugacity to compensate for nonideal gas behavior. The effect of pressure upon the diffusivity has been determined by Shelby to obey the following empirical expression:

$$D = K/[\phi S(1-2\times 10^{-9}P)], \quad (2)$$

where S and K are the ideal or low-pressure solubility and permeability values, and ϕ is the fugacity coefficient at the experimental pressure P (in Pascals). The helium permeation leak described in this paper (denoted NBSL40) had a maximum reservoir pressure of 1 MPa, which from Eq. (2) leads to predicted maximum nonlinearities of 0.2% in the leak rate variation with helium concentration due to gas-gas interactions within the glass interstices (assuming that the glass in NBSL40 has similar properties to the vitreous silica Shelby used). As will be shown below, this nonlinearity is below the random noise in the leak rate measurement and is therefore not a problem here.

NBSL40 was constructed using a commercially available pyrex glass leak element which we then enclosed in a stainless-steel reservoir with all-metal seals. The leak was connected to the leak manifold and the leak reservoir was evacuated with a roughing pump. The reservoir was filled to a predetermined pressure with 99.99% He. The reservoir pressure and temperature were allowed to stabilize for a period of 1 h; after this time the pressure (P) was measured with a quartz-bourdon-tube type gauge. The temperature (T) was also recorded for determination of the reservoir helium concentration (C). Typical reservoir pressures were 0.1 to 1.1 MPa, from which the helium reservoir concentrations were calculated using the ideal gas law (in compatible units):

$$C = (N/V) = P/(RT). \quad (3)$$

Closing the valve to the leak reservoir then prevented changes of the reservoir concentration which could occur due to subsequent temperature fluctuations. Since the leak rate of a helium permeation leak generally takes from 48 to 72 h to stabilize to within 0.1% after filling with helium, NBSL40 was allowed to stabilize for a 3-day period before the leak rate was measured. The technique outlined in Sec. II was employed to determine the leak rates of NBSL40 at six reservoir concentrations varying from $(0.4 \text{ to } 4.6)\times 10^{-4}$ mol/cm^3.

Data for the leak rate dependence on reservoir concentration were analyzed using a least-squares technique to fit an equation of the following form:

$$\text{Leak Rate} = A_1 C, \quad (4)$$

where C is the helium concentration in the reservoir (mol/cm^3), and $A_1(T)$ is a constant (for a constant temperature T) with units of cm^3/s. For the data obtained, fitting to Eq. (4) resulted in a value of A_1 (23 °C) $= 4.174\times 10^{-8}$ cm^3/s. A plot of the residuals of this fit is given in Fig. 4. The three-sigma deviation of the coefficient was 0.2%. The maximum total uncertainty in the measured leak rate was 8.4%.

In summary, sintered leaks have a capability of being used over a wide range of leak rates and can be used with any nonreactive gas. Sintered leak elements, although capable of stabilities within 3% over a length of a year, can also exhibit

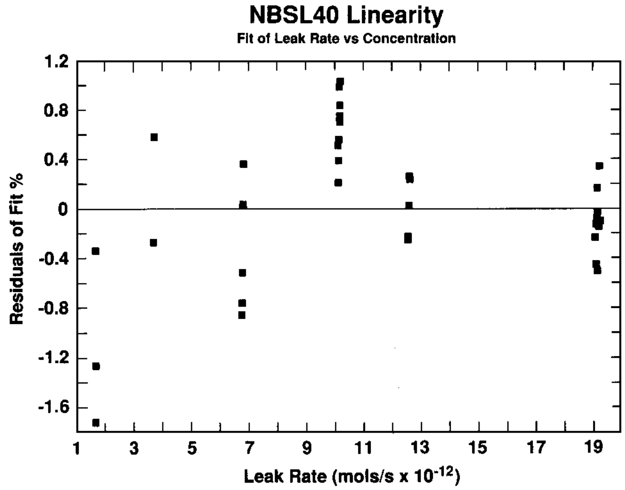

FIG. 4. Residuals of a linear fit of leak rate vs reservoir concentration for variable permeation leak NBSL40.

shifts of 15% over a 3 month period. A permeation VL was tested and the leak rate was determined to be proportional to the helium reservoir concentration. The testing of the permeation VL was conducted over an 8-month period during which time the leak rate was reproducible to within detectable limits when returning to an established reservoir concentration. In order to avoid possible instabilities with a sintered metal leak, and the need to operate leak No. 49 or NBSL5 at unreasonably low reservoir pressures (possibly leading to additional instabilities), a permeation VL (NBSL40) was chosen to be used to determine the linearity of the helium leak detector.

IV. LINEARITY OF THE HELIUM LEAK DETECTOR

Once it had been calibrated, NBSL40 was installed on one port of the leak comparator manifold and a second, fixed-reservoir leak (NBSL10), which had also been calibrated on the Primary Leak Standard, was installed on the second port. NBSL40 was filled to a chosen helium concentration and allowed to stabilize over a 3-day period while being evacuated on the vacuum side with a turbo molecular pump. The leak detector was allowed to warm up for a 24-h period isolated from the leaks. The background ion current of the leak detector was then measured ten times over a 5-min interval using a commercially available digital voltmeter, and the average and standard deviation were calculated. NBSL40 was then valved into the leak detector and the flow rate allowed to stabilize for 60 s before data were collected. Ion current data were collected over a 5-min period, and the average and standard deviation were again calculated based upon the ten readings. NBSL40 was then isolated from the leak detector and NBSL10 was valved into the leak detector and allowed to stabilize for 60 s. Ion current data were collected over a 5-min interval, and the average and standard deviation were calculated. The leak detector was then isolated from both leaks. The indicated leak rate was allowed to stabilize for 60 s and the zeros were recorded following the procedure used for the first step of the process. The average of the "zeros" at the beginning and end was recorded as the zero offset to be used for calculation of the leak rate of NBSL40, as referenced to NBSL10 using the leak detector, for that set of measurements. This process of taking data over a 5-min interval was repeated approximately 100 times for a given NBSL40 reservoir concentration using the automated leak comparator system. The entire procedure as described in this paragraph was repeated for seven reservoir

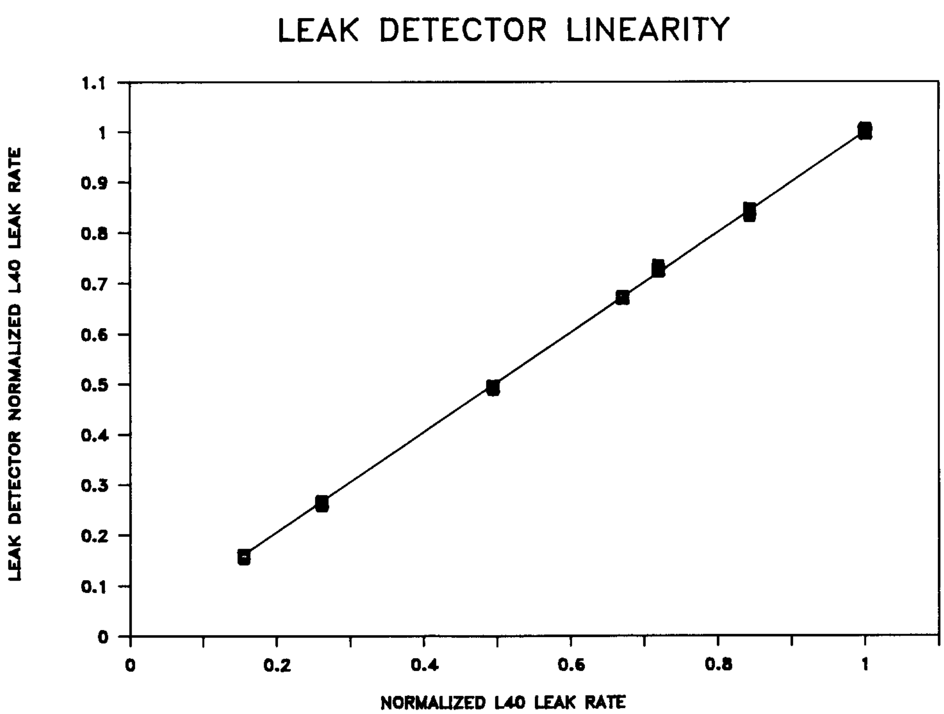

FIG. 5. Normalized leak detector linearity.

concentrations from $(0.5 \text{ to } 2.8)\times10^{-4}$ mol/cm^3, corresponding to leak rates representative of those from the majority of customer leaks. All of these measurements were performed within a single range setting of the leak detector, as is typically the case when determining the temperature dependence of a customer leak.

The characteristics of the leak detector that are important to the interpretation of the data are: the response time of the leak detector is roughly 2 s to reach 63% of the final reading; the minimal detectable leak rate is below 1×10^{-14} mol/s; the ion-current amplifier drift specified by the manufacturer is to be no greater than 1% of full scale per hour.

Instabilities (predominantly drift) of the ion current readings made it impossible to determine accurately the inherent linearity of the leak detector using the uncorrected ion current signal. Zero drift of the ion current has been determined by periodically measuring the ion current with the leak isolated from the leak detector; however, compensating for the drift in gain of the ion current amplifier is more complicated. The drift in the gain was evaluated by periodically valving in a fixed reservoir helium permeation leak (NBSL10), which has a constant leak rate, and measuring the zero-corrected ion current signal of the leak detector. The ion current signal for NBSL40 was then corrected for the drift in the gain of the ion current amplifier, thus determined, and is referred to as the "normalized" data. This enabled us to compare the normalized ion current data (data corrected for zero and gain drift of the ion current amplifier) for NBSL40, as measured by the leak detector, to the known leak rates of NBSL40, as measured on the Primary Leak Standard.

An attempt was made to determine the cause of the instability of the ion current. The emission current was monitored over an extended period of time and fluctuations on the order of 0.1% over a 1-h period were observed. The instability in the ion current reading was isolated to an amplifier that was found to be varying as much as 0.8% of full scale reading per hour. While this was within the manufacturer's specifications previously stated, it leads to significant errors in the measurement of the inherent linearity of the leak detector if corrections are not made.

The measured linearity of the leak detector is presented in Fig. 5. The horizontal axis is the actual leak rate of NBSL40 (as measured on the Primary Leak Standard), plotted as a fraction of the largest value of leak rate used during the leak detector characterization, 1.4×10^{-11} mol/s. The vertical axis is the corresponding value of the leak rate of NBSL4O as measured by the leak comparator, using the technique presented above, expressed at each point as a fraction of 1.4×10^{-11} mol/s. The straight line drawn through the data is the best linear fit for the curve constrained to pass through the point 1,1. If the leak detector were perfectly linear, the slope of the line would be exactly one. The actual slope deviated from unity by 0.39%, with a 3σ uncertainty of 0.024%. The scatter in the data can be seen more clearly in Fig. 6, where the residuals of the data from the straight-line fit in Fig. 5 are plotted against the actual leak rates of NBSL40. The relatively large scatter that characterizes the residuals emphasizes the necessity for averaging, and points out one of the dangers in calibrating a leak detector based on a single measurement.

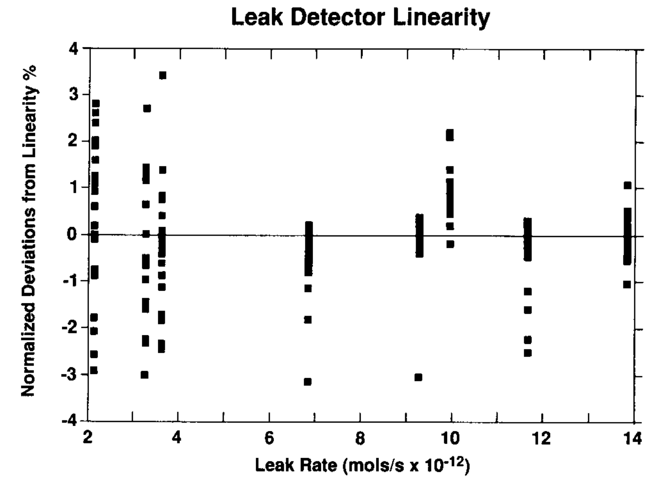

FIG. 6. Leak detector deviation from linearity.

V. SUMMARY

Two types of VLs were examined for their application as transfer standards to determine the linearity of a helium leak detector. Although both the sintered type and helium permeation type leaks covered the desired range of leak rates, the leak rates of the sintered type were found to be nonlinear with reservoir concentration or pressure and these leak rates show instability with time. The helium permeation VL that was characterized in this study (NBSL40) has a leak rate which is directly proportional to the concentration of the helium present in the reservoir for leak rates covering the range 1×10^{-12}–2×10^{-11} mol/s, valid within the uncertainty of the NIST Primary Leak Standard. The 3σ value of the residuals of the linear fit was 0.2%, which indicates that both the permeation leaks and the Primary Leak Standard are highly linear over this range, or that they both deviate in the same, nonlinear manner (which is highly unlikely). Linear behavior of the permeation VL is also predicted by the available theory. The data for this and other permeation leaks also indicate good stability with time. Thus, the permeation VL could be used to evaluate the linearity of the leak detector. The leak detector exhibited an ion current drift rate of the order of 1% of full scale per hour, which would lead to significant errors in leak rate measurement over long periods of time. To countor the effects of the ion current drift, the leak detector zero drift was periodically measured and the drift in the gain was evaluated by periodically measuring the leak rate from a fixed-reservoir leak. Using this technique, the leak detector was determined to be inherently linear to within a fraction of a percent over more than a decade of operation (2×10^{-12}–1.4×10^{-11} mol/s). Since 2–3 days are typically required when measuring temperature coefficients of leaks using the leak detector, the same evaluation and averaging techniques used above are required in order to correct for leak detector random noise and drift, and take advantage of the inherent linearity of the leak detector.

While the technique of determining the linearity of a leak detector with a permeation VL has been used over a range of one decade, it is possible to use this method over a much greater range if the leak rate of the permeation VL were known over a larger range of helium reservoir concentrations. It should be reemphasized that when using permeation VLs, several days are usually required for the leak rate to stabilize after a change in the reservoir concentration has been made. Thus, while the permeation VL offers the flexibility of "infinitely variable" calibrated leak rates using only one leak, a disadvantage is the long period of time required to characterize the leak detector. An effective compromise is to characterize thoroughly the linearity of a leak detector less frequently using a VL, and to use two fixed-reservoir leaks to perform a more routine two-point calibration check.

[a] Guest Researcher from the Precision Instrument Development Center, National Science Council, Taiwan.

[1] K. E. McCulloh, C. R. Tilford, C. D. Ehrlich, and F. G. Long, J. Vac. Sci. Technol. A **5**, 376 (1987).
[2] C. R. Tilford, S. Dittmann, and K. E. McCulloh, J. Vac. Sci. Technol. A **6**, 2853 (1988).
[3] R. W. Hyland, C. D. Ehrlich, C. R. Tilford, and S. Thornberg, J. Vac. Sci. Technol. A **4**, 334 (1986).
[4] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1976).
[5] J. E. Shelby, J. Am. Ceram. Soc. **54**, 125 (1970).
[6] J. E. Shelby, *Treatise On Material Science and Technology* (Academic, New York, 1979), Vol. 17.